Fluctuation Relations for Spintronics

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Fluctuation relations are derived in systems where the spin degree of freedom and magnetic interactions play a crucial role. The form of the nonequilibrium fluctuation theorems relies on the assumption of a local balance condition. We demonstrate that in some cases the presence of magnetic interactions violates this condition. Nevertheless, fluctuation relations can be obtained from the microreversibility principle sustained only at equilibrium as a symmetry of the cumulant generating function for spin currents. We illustrate the spintronic fluctuation relations for a quantum dot coupled to partially polarized helical edge states.

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Introduction.—Nonequilibrium fluctuation theorems (FTs) [1–3], widely used for macroscopic systems, are based on the thermodynamics governing the physical processes when they evolve forward and backward in time. The boundary conditions for the forward and the time-reversed processes determine the balance condition for the entropy exchange and, therefore, the form of the fluctuation theorem [3]. The applicability of the nonequilibrium FTs to quantum systems has become an exciting problem, especially to the case of the charge transfer phenomena in mesoscopic systems in the context of the full counting statistics [4–7]. Interestingly, relations akin to the fluctuation-dissipation theorem [8–11] have been formulated beyond the linear response regime [7,12–22]. These fluctuation relations relate nonequilibrium fluctuation and dissipation coefficients for phase-coherent conductors. However, the role of a genuine quantum property such as the spin degree of freedom in the fluctuation relations has not yet been investigated in detail. Our motivation is not only fundamental since the electronic spin offers enormous advantages to create devices with unusual and extraordinary new functionalities [23]. The purpose of this work is thus to generalize the fluctuation relations for spintronic systems.

Fluctuation relations are generated from the cumulant generating function (CGF) $\mathcal{F}(\chi) = \ln\sum_Q P(Q,t)e^{-i\chi Q}$, where $P(Q)$ is the charge distribution function. Firstly, the CGF $\mathcal{F}$ is expanded in a Taylor expansion in terms of affinities $A = (qV_1/k_BT, qV_2/k_BT, \ldots)$ (q is the electron charge, k_B is the Boltzman constant, T is the temperature, and V_i, $i = 1, 2\ldots$ are the applied voltages) and counting fields $\chi = (\chi_1, \chi_2, \ldots)$ around the equilibrium condition. Then, thanks to the symmetries $F(0, A) = 0$ (charge probability conservation condition) and $F(-A, A) = 0$ (global detailed balance condition), fluctuation relations among the higher-order nonlinear cumulants are found. Indeed, the symmetries of $\mathcal{F}$ can be considered as the nonequilibrium FT versions for the currents within a transport theory. Initial experiments by using a mesoscopic dot interferometer have tested these relations [24]. In this experiment, the noise susceptibility and the second-order conductance were found to be proportionally related.

Spins are sensitive to magnetic fields and also to electric fields due to spin-orbit interactions. Fluctuation relations for the charge transport have been formulated in the presence of magnetic fields [14–16]. In Ref. [14], the nonequilibrium FT for the forward and backward charge distribution probability at opposite B polarities $P(Q,B)/P(-Q,-B) = e^{QA}$ was used to derive such fluctuation relations. However, some caution is needed since $P(Q,B)$ and $P(-Q,-B)$ are considered for a system driven out of equilibrium in which the interacting internal potentials are no longer even functions of B [25] and the application of such theorem may break down [15]. To circumvent this obstacle, Ref. [15] uses a symmetry of $\mathcal{F}$ associated with the microreversibility condition only at equilibrium $P(Q,B)_{A\to 0} = P(-Q,-B)_{A\to 0}$. We here derive the spintronic fluctuation relations in the same spirit when time-reversal symmetry is broken not only by external magnetic fields but also by the presence of ferromagnetic electrodes. In this case, at equilibrium $P(Q,B,p)_{A\to 0} = P(Q,-B,-p)_{A\to 0}$, where p is the lead magnetization [26].

We illustrate our findings with a quasilocalized level coupled to helical edge states that are partially polarized by the presence of polarized electrodes [see Fig. 1(b)]. Helical modes have been observed in topological insulators [27] and proposed to occur in quantum wires [28] and in carbon nanotubes [29]. This quantum spin Hall state consists of gapless excitations that exist at the boundaries in which its propagation direction is correlated with its spin due to the spin-orbit interaction. By electrostatic gating, quasilocalized states can form in the interior of the carbon nanotubes and quantum wires. Furthermore, ferromagnetic contacts have been successfully attached to these nanodevices [30]. Finally, Ref. [31] suggests creating a quasibound state in quantum spin Hall setups by using ferromagnetic insulators that serve as tunneling barriers.

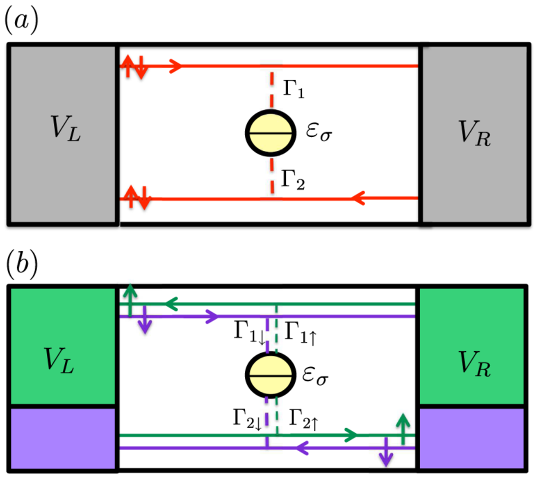

FIG. 1 (color online). (a) Sketch of a quasilocalized level (ε_σ) coupled to chiral edge states with $\Gamma_{1,2}$ and driven out of equilibrium with V_L and V_R bias voltages. For $B > 0$ the upper (lower) edge state is injected from V_L (V_R). (b) Localized level coupled to unequally spin populated helical edge states due to the spin injection from the ferromagnetic leads. Then, the tunneling couplings are spin-dependent: $\Gamma_{(1,2)\uparrow(\downarrow)}$. Ferromagnetic electrodes: larger light area (green) corresponds to majority spins whereas smaller dark area (purple) are minority spins. Helical states: upper left (upper right) movers are spin up (spin down) carriers injected from V_R (V_L).

Local detailed balance.—Consider a system described by a set of m discrete states coupled to ℓ electronic reservoirs. We assume that its dynamics is governed by the master equation $d\rho/dt = \mathcal{W}\rho$, where $\mathcal{W}$ is the transition rate matrix, and ρ denotes the occupation probabilities for the m states. The exchange of energy ($\Delta E^{(\ell)}$) or particles ($\Delta N^{(\ell)}$) in the ℓth reservoir with inverse temperature $\beta^{(\ell)}$ is described by adding counting fields ($\chi_E^{(\ell)}$, $\chi_N^{(\ell)}$) to the off-diagonal matrix elements of $\mathcal{W}$. Thus, for the upper off-diagonal, the transition rate from the state n to the state m are modified according to $W_{nm} = \sum_\ell W_{nm}^{(\ell)} e^{\chi_E^{(\ell)}\Delta E^{(\ell)} + \chi_N^{(\ell)}\Delta N^{(\ell)}}$ (for $n < m$), whereas for the lower off-diagonal terms these rates are $W_{nm} = \sum_\ell W_{nm}^{(\ell)} e^{-\chi_E^{(\ell)}\Delta E^{(\ell)} - \chi_N^{(\ell)}\Delta N^{(\ell)}}$ ($n > m$). Usually, boundary conditions are taken into account through the local detailed balance (LDB) condition in which weight factors $e^{-\beta^{(\ell)}(\mathcal{H}_\ell - \mu N_\ell)}$ ($\mathcal{H}_\ell$, and N_ℓ denote the Hamiltonian and the particle number operator, respectively, for the ℓth reservoir) balance forward and backward processes. To be more specific,

$$\frac{W_{nm}^{(\ell)}}{W_{mn}^{(\ell)}} = e^{-\beta^{(\ell)}(\Delta E^{(\ell)} - \mu^{(\ell)}\Delta N^{(\ell)})}. \tag{1}$$

From the LDB condition, the equality $\mathcal{W}(\chi_E^{(\ell)}, \chi_N^{(\ell)}) = \mathcal{W}^T(\beta^{(\ell)} - \chi_E^{(\ell)}, -\beta^{(\ell)}\mu^{(\ell)} - \chi_N^{(\ell)})$ is automatically satisfied, reflecting the following symmetry for the generating function $\mathcal{F}$ [which is constructed from $\mathcal{W}(\chi_E^{(\ell)}, \chi_N^{(\ell)})$]:

$$\mathcal{F}[\chi_E^{(\ell)}, \chi_N^{(\ell)})] = \mathcal{F}[\beta^{(\ell)} - \chi_E^{(\ell)}, -\beta^{(\ell)}\mu^{(\ell)} - \chi_N^{(\ell)}]. \tag{2}$$

Although in many systems we can assume some type of LDB condition, in general Eq. (1) is not fulfilled [32]. To see this in a quantum conductor, we consider the system sketched in Fig. 1(a) in which the presence of a magnetic field breaks time-reversal symmetry. The system consists of a quasilocalized state with energy ε_d in the Coulomb blockade regime coupled to two chiral states propagating along the opposite edges of a quantum Hall conductor (filling factor $\nu = 1$) [16,25,33]. In the infinite charging energy limit case, only two dot charge states are permitted: $|0\rangle$ and $|1\rangle$. For positive magnetic fields $B > 0$, carriers in the upper (lower) edge state move from the left (right) terminal to the right (left) terminal. The current flow is reversed for $B < 0$. Interaction between the quasilocalized state and the edge states takes place via tunnel couplings Γ_1 and Γ_2 and capacitive couplings C_1 and C_2. The chiral coupling involves different transition rates depending on the polarity of the magnetic field. For a positive B, we have $W_{01}^{L(R)} = \Gamma_{1(2)} f(B, \mu_{L(R)})$, $W_{10}^{L(R)} = \Gamma_{1(2)}[1 - f(B, \mu_{L(R)})]$, where $f(B, \mu_{L(R)}) = 1/(1 + \exp\beta[\mu_d(B) - \mu_{L(R)}])$ is the Fermi-Dirac distribution function, $\mu_{L(R)} = qV_{L(R)} + E_F$ denotes the electrochemical potential in the lead $L(R)$ with $E_F = 0$ as the Fermi energy, and μ_d is the electrochemical potential of the quasilocalized state which is self-consistently calculated and depends on the B orientation [16]. For $B > 0$, $\mu_d(B) - \mu_L = \varepsilon_d - (1-\eta)qV/2$, where $\eta = (C_1 - C_2)/(C_1 + C_2)$ is the capacitance asymmetry parameter and $V = V_L - V_R$. For $B < 0$, the motion of the edge states is reversed and then $\mu_d(-B) - \mu_L = \varepsilon_d - (1 + \eta)qV/2$. Because of the fact that $\mu_d(B) \neq \mu_d(-B)$, the LDB condition is not satisfied. Clearly,

$$\frac{W_{10}^L(B)}{W_{01}^L(-B)} = e^{\beta(\varepsilon_d - qV/2)}\left[1 - \beta\eta\frac{qV}{2}\tilde{f}_{eq} + \mathcal{O}(V^2)\right], \tag{3}$$

where β is the common inverse temperature and $\tilde{f}_{\rm eq} = 1 - 2f_{\rm eq}$ with $f_{\rm eq}$ being the Fermi function at equilibrium ($V_L = V_R$). Importantly, the violation of the LDB condition occurs for asymmetric capacitance couplings only. In the symmetric case or at equilibrium, Eq. (1) is recovered. The violations of LDB are thus a consequence of asymmetric, chiral states out of equilibrium.

We now show that violations of the LDB conditions are also present in the absence of magnetic fields and when the spin degree of freedom is explicitly accounted for. For that purpose, we consider the system sketched in Fig. 1(b) a quasibound state that is tunnel-coupled to helical edge states. The helical modes are partially polarized due to their coupling to two ferromagnetic electrodes with parallel magnetization and equal polarization p. In this manner, polarized helical edge states are described with a spin-dependent density of states (DOS) $D_{is} = (1 + sp)D_i/2$, where $s = +(-)$ for $\uparrow$($\downarrow$)-helical mode and, D_i, with $i = u, d$ denoting the upper and lower edge state DOS in the absence of polarization [34]. In the wide-band limit approximation, the tunneling rates become spin-dependent, $\Gamma_{is} = \pi|t_i|^2 D_{is}$, with $|t_i|^2$ the tunnel probability from the ith edge state. Defining $\Gamma_i = \pi|t_i|^2 D_i$, we find $\Gamma_{is} = (1 + sp)\Gamma_i/2$.

Our transport description also includes an electrostatic model for interactions between the dot and the edge states. Within the mean-field approach, the electrochemical capacitive coupling C_μ consists of a geometrical capacitance contribution, C_g, which depends on the width and the height of the tunnel barrier, and a quantum capacitance term, $C_{is,q}$, which we take as proportional to D_{is}:

$$C_{\mu is}^{-1} = \frac{1}{C_g} + \frac{1}{q^2 D_{is}}. \tag{4}$$

We emphasize that the capacitive couplings are, in general, spin-dependent [35]. For sufficiently large geometrical capacitances, $C_g \gg C_{is,q}$, we find from Eq. (4) the capacitative couplings

$$C_{u1(2)} = \frac{1+p}{2} C_{1(2)}, \qquad C_{u3(4)} = \frac{1-p}{2} C_{3(4)}, \tag{5}$$

where $C_i = q^2 D_i$ with $C_{u1(d1)}$, $C_{u2(d2)}$ being the capacitive couplings between left (right) movers with up (down) spin along the top edge and the dot electron with spin $\uparrow$ ($\downarrow$), whereas $C_{u3(d3)}$ and $C_{u4(d4)}$ couple the same dot state with right (left) movers along the bottom edge with up (down) spins (see Fig. 2). For thin edge states, D_i depends on the steep confinement potential at the top and bottom edges, which will generally differ [34]. Then, we take $C_1 = C_3$ and $C_2 = C_4$ but $C_1 \neq C_2$. As a consequence, the capacitive couplings between the dot and the edges is asymmetric: $\eta \neq 0$. Furthermore, since the upper and lower edge modes are equally polarized, one has $C_{dj} = C_{uj}$.

Consider for the moment the case where the capacitance coupling between the dot states is neglected ($C = 0$). Then, we calculate the spin-dependent electrochemical

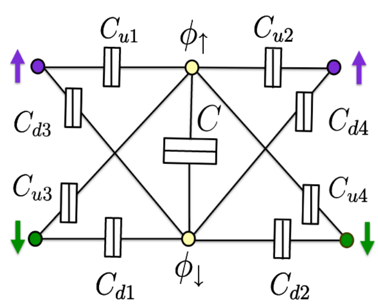

FIG. 2 (color online). Electrostatic model for the quasilocalized state connected to helical spin edge states. Spin up(down) localized level is capacitatively coupled to the upper up (down) helical channel with capacitances $C_{u1(u3)}[C_{d1(d3)}]$ and to the lower up(down) helical channel with capacitances $C_{u2(u4)}[C_{d2(d4)}]$. A mutual capacitance between up and down localized levels is accounted for with C. $\phi_{\uparrow(\downarrow)}$ denotes the spin up(down) internal potential for the quasibound state.

potential of the dot and find the simple relation $\mu_\sigma(p) - \mu_{\bar\sigma}(-p) = p\eta V$. Now, in a time-reversal operation, we have to invert the lead polarization, the edge state spin index, and the dot spin. Doing so, we obtain an invariant result only at equilibrium ($V = 0$) or for symmetric capacitive couplings. But, in general, when $V \neq 0$ the original state is not restored and, as a consequence, LDB is not fulfilled:

$$\frac{W_{0\sigma}(p)}{W_{\bar\sigma 0}(-p)} = e^{\beta[\varepsilon_d - qV/2]}\left[1 - \beta\eta p\frac{qV}{2}\tilde f_{eq} + \mathcal{O}(V^2)\right], \tag{6}$$

where $\sigma = \{\uparrow, \downarrow\}$ denotes the dot spin index. We stress that helicity is needed in our example to find departures from LDB. Although we cannot rule out the possibility that nonchiral, spintronic systems (e.g., a dot directly attached to ferromagnetic leads) might show such departures if coherent tunneling or strong correlations are taken into account, our conceptually simple system already exhibits the effect with fully analytical expressions.

Spintronic fluctuation relations.—We now treat on equal footing the presence of both magnetic fields and polarized contacts. The spin-dependent probability distribution satisfies the microreversibility condition, but only at equilibrium

$$P(\{n_{\alpha s}, n_{\beta s'}, \ldots\}; B, p) = P(\{-n_{\alpha\bar s}, -n_{\beta\bar s'}, \ldots\}; -B, -p), \tag{7}$$

where α and s are the lead and spin indices, respectively, and $p \equiv (p, p', \ldots)$ contains the magnetizations for the leads. The CGF $\mathcal{F}(\{i\chi\}, A)$ can be expanded in terms of powers of voltages and counting fields

$$\mathcal{F}(\{i\chi\}, A) = \sum_{\{k_{\alpha s}\},\{l_\alpha\}} f_{\{k_{\alpha s}\},\{l_\alpha\}} \frac{\prod_{\alpha s}(i\chi_{\alpha s})^{k_{\alpha s}} \prod_\alpha A_\alpha^{l_\alpha}}{\prod_\alpha k_{\alpha s}! \prod_\alpha l_\alpha!} \tag{8}$$

and

$$f_{\{k_{\alpha s}\},\{l_\alpha\}} = \prod_{\alpha s} \partial^{k_{\alpha s}+l_\alpha} \mathcal{F}(\{i\chi\}, A)/\partial(i\chi_{\alpha s})^{k_{\alpha s}} \partial A_\alpha^{l_\alpha}|_{i\chi\to 0, A\to 0}, \tag{9}$$

where k and l are nonnegative integers. From the derivatives of $\mathcal{F}(\{i\chi\}, A)$ with respect to the counting fields, the cumulants are generated. In this way, the average current through terminal α with spin s is derived from Eq. (8) as $\langle I_{\alpha s}\rangle = f_{\{1_{\alpha s}\}}$. Similarly, second cumulant (current-current correlation) $S_{\alpha s,\beta s'} \equiv \langle \Delta I_{\alpha s} \Delta I_{\beta s'}\rangle$ ($\Delta I_{\alpha s} = \hat I_{\alpha s} - \langle I_{\alpha s}\rangle$), where $\hat I$ denotes the current operator) and the third cumulant $C_{\alpha s\beta s'\gamma s''} \equiv \langle \Delta I_{\alpha s}\Delta I_{\beta s'}\Delta I_{\gamma s''}\rangle$ are given by $S_{\alpha s\beta s'} = f_{\{1_{\alpha s}1_{\beta s'}\}}$ and $C_{\alpha s\beta s'\gamma s''} = f_{\{1_{\alpha s}1_{\beta s'}1_{\gamma s''}\}}$, respectively. We expand both the current $\langle I_{\alpha s}\rangle$ and the noise $\langle S_{\alpha s\beta s'}\rangle$ in powers of the applied voltages as follows:

$$\langle I_{\alpha s}\rangle = \sum_j G^{(1)}_{\alpha s,j} V_j + \frac{1}{2}\sum_{j,k} G^{(2)}_{\alpha s,jk} V_j V_k + \mathcal{O}(V^3),$$

$$\langle S_{\alpha s\beta s'}\rangle = S^{(0)}_{\alpha s\beta s'} + \sum_j S^{(1)}_{\alpha s\beta s',j} V_j + \mathcal{O}(V^2). \tag{10}$$

Here $G^{(1)}_{\alpha s,j} = f_{\{1_{\alpha s}\},\{1_j\}}$ corresponds to the linear conductance, $G^{(2)}_{\alpha s,jk} = f_{\{1_{\alpha s}\},\{1_j 1_k\}}$ is the second-order conductance, and $S^{(1)}_{\alpha s,\beta s',j} = f_{\{1_{\alpha s} 1_{\beta s'}\},\{1_j\}}$ is the noise susceptibility. Fluctuation relations are expressions that relate the f coefficients at different order in voltage. To derive explicitly these relations, we employ the microreversibility condition at equilibrium $F(i\chi_{\alpha s}, i\chi_{\beta s'}, \cdots, A, +B)|_{A=0} = F(-i\chi_{\alpha\bar{s}}, -i\chi_{\beta\bar{s}'}, \cdots, A, -B)|_{A=0}$ [cf. Eq. (7)]. It is convenient to define the symmetrized (+) and antisymmetrized (−) combination of the f factors

$$f^{\pm}_{\{k_{\alpha s}\},\{l_j\}} = f_{\{k_{\alpha s}\},\{l_j\}}(B,p) \pm f_{\{k_{\alpha\bar{s}}\},\{l_j\}}(-B,-p), \tag{11}$$

where $f_{\{k_{\alpha\bar{s}}\},\{l_j\}}(-B,-p)$ is generated by means of time-reversal operation $B \rightarrow -B$, $p \rightarrow -p$, and $s \rightarrow -s$. According to Eq. (7), the $f^{\pm}$ factors are even(odd) functions under time-reversal operation. This even-odd property is translated into the following relations for the equilibrium coefficients [in the sense of a voltage expansion, see Eqs. (10)]:

$$S^{(0)}_{\alpha s\beta s'}(B,p) = S^{(0)}_{\alpha\bar{s}\beta\bar{s}'}(-B,-p),$$

$$C^{(0)}_{\alpha s\beta s'\gamma s''}(B,p) = -C^{(0)}_{\alpha\bar{s}\beta\bar{s}'\gamma\bar{s}''}(-B,-p). \tag{12}$$

Now by using the global detailed balance condition $\mathcal{F}(-A,A)_{\pm} = 0$, and the probability conservation $\mathcal{F}(0,A)_{\pm} = 0$, one can derive the spintronic fluctuation relations among different $f_{\pm}$ factors. Here we explicitly show those that relate the coefficients appearing in the third cumulant, noise, and the conductances in the voltage expansion of Eq. (10):

$$C^{(0)}_{\alpha s\beta s'\gamma s''\pm} = k_B T[S^{(1)}_{\alpha s\beta s',\gamma\pm} + S^{(1)}_{\alpha s\gamma s'',\beta\pm} + S^{(1)}_{\beta s'\gamma s'',\alpha\pm} - k_B T(G^{(2)}_{\alpha s,\beta\gamma\pm} + G^{(2)}_{\beta s',\alpha\gamma\pm} + G^{(2)}_{\gamma s'',\alpha\beta\pm})]. \tag{13}$$

Fluctuation relations between even higher-order response coefficients toward the strongly nonequilibrium domain can be similarly found, relating different current cumulants at different order; however, the resulting expressions, already in the spinless case, look rather cumbersome [15].

We verify Eq. (13) in a multiterminal setup in which the LDB condition is broken. For that purpose we generalize the two terminal quantum-spin Hall bar system [Fig. 1(b)] to the multiterminal case in which upper and lower helical modes are now connected to different terminals V_i, $i = 1 \cdots 4$ [see inset in Fig. 3(d)]. We additionally consider spin-flip relaxation events within the quasibound state that can occur due to spin-spin interactions with a spin fluctuating environment (hyperfine interaction, spin-orbit interactions, etc.). We phenomenologically model this rate as $\gamma^{\sigma\bar{\sigma}}_{sf} = \gamma_{sf}\exp[(\varepsilon_\sigma - \varepsilon_{\bar{\sigma}})/(2k_B T)]$. Notice that due to spin-flip events, spin up and down currents are correlated and then Eq. (13) is satisfied in a nontrivial manner. We emphasize that Eq. (13) is verified (see Fig. 3) even for a finite capacitance asymmetry where the LDB condition is not met.

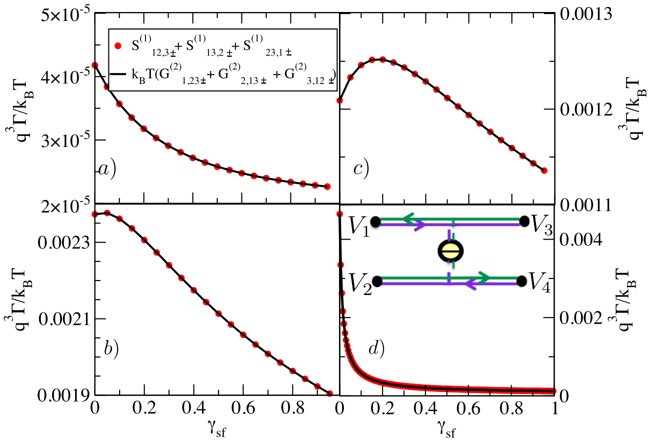

FIG. 3 (color online). Verification of spintronic fluctuation relations as a function of γ_{sf} in the presence of magnetic interactions, B, and p for different values of polarization p: (a) $p = 0$, (b) $p = 0.25$, (c) $p = 0.5$, and (d) $p = 0.75$. Upper helical modes: left (right) movers are spin up (down) injected with voltage $V_{1(3)}$. Lower helical modes: right (left) movers are spin up (down) injected with voltage $V_{2(4)}$. Parameters: $\Gamma = 1$, $q^2/[4(C_1 + C_2)] = 40\Gamma$, $\epsilon_d = 0$, $k_B T = 5\Gamma$, $g\mu_B B = 0.1\Gamma$, and capacitance asymmetry $\eta = 0.5$. Note that in our chiral system, spin indices are included in the lead indices for the fluctuation relations.

Conclusions.—In short, we have shown that the applicability of nonequilibrium FT when magnetic interactions are present is not *a priori* ensured. We illustrate this statement by using a quasilocalized level coupled to a chiral one-dimensional conducting channel. We demonstrate that local detailed balance condition is not satisfied when a magnetic field is included and the system is driven out of equilibrium. Importantly, we have derived the fluctuation relations for spintronic systems and have explicitly verified them in the illustrative case of a quasilocalized state coupled to partially polarized helical edge states. Our formalism is based on zero-frequency fluctuations and time-independent fields but in the presence of arbitrary interactions. Promising avenues for future work include finite-frequency calculations and ac fields.

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